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Abstract

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The long-term assessment of air quality on an island in Malaysia

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Abstract

This study aims to evaluate the air quality on Langkawi Island, a famous tourist destination in Malaysia, using 13 years of data (1999–2011) recorded by the Malaysian Department of Environment. Variations of seven air pollutants (O_3 , CO, NO, NO_2 , NO_x , SO_2 and PM_{10}) and three meteorological factors (temperature, humidity and wind speed) were analysed. Statistical methods used to analyse the data included principal component regression (PCR) and sensitivity analysis. The results showed PM_{10} was the dominant air pollutant in Langkawi and values ranged between $5.0 \mu g m^{-3}$ and $183.2 \mu g m^{-3}$. The patterns of monthly values showed that the concentrations of measured air pollutants on Langkawi were higher during the south-west monsoon (June–September) due to seasonal biomass burning activities. High CO/NO_x ratio values (between 28.3 and 43.6), low SO_2/NO_x ratio values (between 0.04 and 0.12) and NO/NO_2 ratio values exceeding 2.2 indicate the source of air

pollutants in this area was motor vehicles. PCR analysis grouped the seven variables into two factor components: the F1 component consisted of SO₂, NO and NO_x and the F2 component consisted of PM₁₀. The F1 component ($R^2 = 0.931$) indicated a stronger standardized coefficient value for meteorological variables compared to the F2 component ($R^2 = 0.059$). The meteorological variables were statistically significant ($p < 0.05$) in influencing the distribution of the air pollutants. The status of air quality on the island could be improved through control on motor vehicle emissions as well as collaborative efforts to reduce regional air pollution, especially from biomass burning.

Keywords: Environmental science, Atmospheric science

1. Introduction

Air pollution occurs when there are high levels of contamination by pollutants in the atmosphere. This can be life threatening and can disrupt the stability of the ecosystem. Modern nations describe air pollution as a serious environmental problem affecting life expectancy, especially in developing countries [1, 2]. Increasing evidence from research undertaken in recent years supports the influence of air pollution on health issues such as lung cancer, respiratory infections, cardiovascular diseases, degenerative brain diseases and high morbidity risks [3, 4, 5, 6, 7, 8, 9, 10]. Due to the adverse effects on human health, air pollution is classified as one of the environmental problems that has become a major global concern [11, 12, 13].

Air pollution in Malaysia generally originates from three major sources: stationary sources, open burning sources and mobile sources [14, 15]. Stationary sources in Malaysia include the emission of dust from urban construction works and quarries, energy power plants, incinerators and industries [16]. Open burning sources include local pollution from vegetation burning emissions and trans-boundary pollution, especially during forest fires caused by peat combustion from neighbouring countries [17]. Mobile sources refers to traffic emissions. In 1998, there were 7.7 million motor vehicles registered with the Road Transport Department of Malaysia. The numbers have since been increasing from year to year and there were 10.6 million motor vehicles registered in 2001. Urbanized areas in Malaysia were the main contributors to the rise of motor vehicles on the road. Kuala Lumpur, located in the Klang Valley, recorded the highest number of motor vehicle registered [18] which exposed the area to high emissions of air pollutants [15, 19, 20]. Severe traffic congestion has the potential to increase emissions of pollutants and degrade the air quality of the area [15, 21, 22, 23]. One of the factors that is related to traffic congestion in an area is the development of tourism [24]. An island will commonly become an area for the development of tourism since tourism is prominent in coastal areas around the world [25]. Severe traffic congestion will then be a problem

plaguing the island with the rise in tourist arrivals contributing to high numbers of motor vehicles on the road affecting the air quality.

Langkawi is one of the developed islands in Malaysia that has received increasing tourist arrivals every year, especially after the declaration of Langkawi Island as a tax-free zone to improve the economy of Malaysia [26]. Due to this tax-free zone status, Langkawi Island has turned into a famous tourist island and has undergone rapid development for the tourism sector. In order to fulfil the demands of the tourism sector, which can contribute significantly to the Gross Domestic Product (GDP) of Malaysia, infrastructure such as road networks on Langkawi Island have been developed. Measures were also taken to enhance facilities to attract more tourists to visit Langkawi Island [27, 28]. The rise in tourist arrivals, especially during school breaks and public holidays, has resulted in an increase in traffic congestion on the island. The tourist arrivals recorded increased from 209,763 in 1986 to 1,835,245 in 2005 and continued to increase to 2,300,000 in 2007 [29, 30].

A previous study by Latif et al. [31] showed that the air quality of Langkawi Island was influenced by motor vehicle traffic and anthropogenic activities such as the cement industry. Langkawi Island is also affected by particulate matter from biomass burning in Sumatra during the Southwest monsoon (June to September) and in Indo-China during the Northeast monsoon (January to March) that occurs almost every year. Besides, due to the island's location close to Sumatra, Indonesia, Langkawi often experiences haze episodes caused by open burning problems [32, 33]. Other than that, air pollutants on Langkawi Island are expected to come from local burning, commercial activities, road dust, construction activities, port activities as well as natural contributions such as fugitive dust and sea spray emissions. The sources are common sources of air pollutants in Malaysia and other tropical countries [34, 35].

This study focuses on the assessment of air quality on Langkawi Island for a 13-year period (1999–2011) to determine the temporal trends of air pollutant concentrations on the island. Application of the Hierarchical Agglomerative Cluster analysis (HACA) in this study was important to determine diurnal air quality patterns of measured air pollutants by clustering homogenous air pollutants while data analysis using box plot interpretation to determine the seasonal and long term annual trends. In addition, this study attempts to estimate the air pollutant sources and meteorological parameters that influence air quality on Langkawi Island. The use of Principal Component Regression (PCR) and sensitivity analysis provides information on the most significant air pollutants and meteorological parameters contributing towards temporal variation in Langkawi Island. Ratio of main criteria air pollutants has been used to determine potential sources of air pollutants. This study does not use any source apportionment analysis based on the composition of major air pollutant such as particulate matter to predict sources of air pollutants in this island.

2. Materials and methods

2.1. Location of sampling station

Langkawi is an island located off the north-west coast of Peninsular Malaysia, in the Andaman Sea region. The island, a tourism location in Malaysia, has an area of approximately 479 km². In 2010, with a population of around 92,893, the island of Langkawi was covered by forested mountains, hills, limestone structures and beaches [36, 37]. Langkawi Island has one continuous air quality monitoring station which is managed by Alam Sekitar Malaysia (ASMA). The location of the air quality monitoring station is at the Langkawi Sports Complex with coordinates N06° 19.903', E099° 51.517' (Fig. 1). The Langkawi air quality monitoring station is about 1.5 km from Kuah, the city centre of Langkawi. This station is categorized as a suburban monitoring station by the Department of Environment (DOE) Malaysia and is surrounded by residential areas, hotels and forests [38].

2.2. Data collection

The observation data used for the analysis period was obtained from the Air Quality Division of the DOE; the Ministry of Natural Resources and Environment of Malaysia. This is a thirteen-year dataset, starting from January 1999 to December 2011. The air quality dataset consisted of ten variables that can be divided into two groups: major air pollutants and meteorological parameters. The major air pollutant group includes ground level ozone (O₃), carbon monoxide (CO), sulphur dioxide (SO₂), nitrogen oxide (NO), nitrogen dioxide (NO₂), oxides of nitrogen (NO_x), and particulate matter with a diameter size of less than 10 µm (PM₁₀) while the meteorological parameter group consisted of wind speed, ambient temperature and relative humidity.

The gas pollutants (O₃, CO, NO, NO₂, NO_x and SO₂) were measured hourly using a Teledyne API instrument (Teledyne Technologies Inc., USA). The O₃ and CO

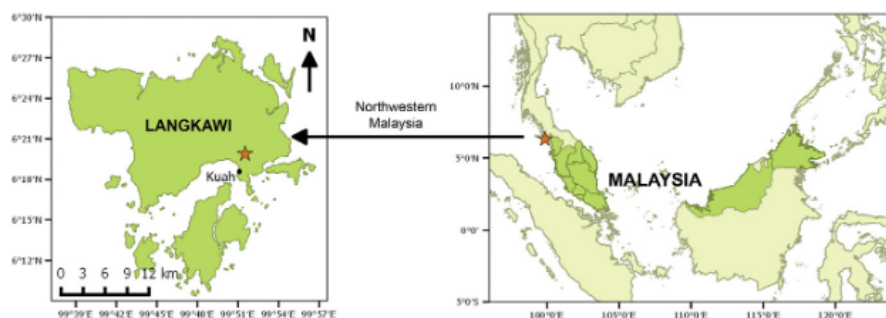


Fig. 1. Location of Langkawi Island in Malaysia, which located at the northwestern of Peninsular Malaysia. The star symbol indicates the air quality monitoring station in Langkawi Island. The black dot symbol refers to Kuah, the city centre of Langkawi Island that located 1.5 km from the Langkawi air quality monitoring station.

concentrations were measured using a Teledyne API Model 400/400E and Teledyne API Model 300/300E, respectively. Both instruments applied the Beer Lambert principle in their detection of O₃ and CO concentration. NO, NO₂ and NO_x were measured by a Teledyne API Model 200A/200E which uses chemiluminescence to detect the chemical compounds. The concentration of SO₂ was measured using a Teledyne API Model 100A/E which applies the UV fluorescence principle. The detection limit for the Teledyne API Model 400A/E, 200A/E and 100A/E is 0.4 ppb, while the detection limit of the Teledyne API Model 300A/E is 0.04 ppm. The concentration of PM₁₀ was measured over 24 h using a Beta Attenuation Mass Monitor 1020 (BAM-1020) (Met One Instrument, Inc., USA). This instrument can detect concentrations of PM₁₀ of less than 4.8 µg m⁻³ (1 h) and less than 1.0 µg m⁻³ (24 h). The ambient temperature was measured by a Met One 062 sensor and relative humidity was measured by a Met One 083D sensor. The wind speed and wind direction were measured using a Met One 010C sensor and a Met One 020C sensor respectively [39].

2.3. Statistical analysis

The univariate statistical descriptive analysis was performed to determine the minimum, maximum, first quartile (Q1), median, third quartile (Q3), mean and standard deviation, analysing one variable at time. The analysis was applied to all the hourly averages of the year concentrations for the main air pollutant group, except for PM₁₀, which is based on daily average (24 h) concentrations. Pollutant concentrations were compared with the maximum allowable values detailed by the Recommended Malaysian Air Quality Guidelines (RMAQG) and the World Health Organization (WHO). These basic statistical values are important when explaining the distribution of the variables from 1999 to 2011 since Langkawi Island was undergoing the process of urbanization. Besides, the result is useful in determining the air pollution status of the island.

The Hierarchical Agglomerative Cluster analysis (HACA) was applied to the diurnal data sets to study the temporal patterns of measured air pollutants. HACA was run using the XLSTAT 2014 version 8.2.00 software. The primary purpose of the HACA analysis is to assemble objects based on the characteristics they possess. In this study the analysis was performed using the Ward's method that utilizes Euclidean distance as a measure of similarity. According to McKenna Jr [40], the temporal patterns of air quality can be determined by arranging all the variables into clusters showing within-group homogeneity and between-group heterogeneity. This common technique will produce several numbers of clusters that can be presented in the form of chart called a dendrogram or as a hierarchical tree. A dendrogram can be defined as one type of graphical representation of cophenetic matrix; either by comparing the cophenetic similarity or cophenetic distance [41]. The

two clusters having the minimum Euclidean distance are then merged; thereby the number of clusters is reduced by one. This procedure is repeated until the whole data set is merged to form a single cluster [42].

2.4. Sensitivity analysis

The sensitivity analysis used in this study is One-factor-at-a-time (OAT) regression. In this study, the OAT regression analysis technique was used to study the level of importance of each input (CO, NO, NO₂, NO_x, O₃, PM₁₀, and SO₂) on the output (vehicles). OAT regression analysis is designed to examine the importance level or significance of the variables concerned [43]. The factor is varied until the best setting of the factor is found but only one factor can vary at a time, so the effects of factor interactions and multi-response interdependences are ignored [44]. According to Shi et al. [45], this analysis will quantify the contribution of each parameter to forecast the level of pollution by the variability of the dependent variable and independent variables. The correlation of determination, (R^2) is a correlation coefficient resulting from the regression analysis.

2.5. PCR and ratio analysis

Principal component regression (PCR) analysis is a combination of PCA (principal component analysis) and OLS (Ordinary Least Squares) regression. According to Sousa et al. [46], PCR analysis has the ability to reduce the multicollinearity in datasets. The presence of multicollinearity among the independent variables will produce invalid results in terms of the model's predictions and determination of the significant independent variables. PCA, one part of the combination in PCR analysis, is a method that reduces the dataset dimensionality by performing a covariance analysis between the factors. The PCA will maximize the correlation between the original variables and new uncorrelated variables that are mutually orthogonal [47]. The factors with significant eigenvalues of more than 1.0 are chosen in order to fully understand the correlation relationship between the variables as the factors are considered significant. PCA was used to filter the data in order to obtain only significant independent variables that are responsible for the air pollutant concentrations (dependent variables) observed could be determined. Then, the significant factors, consisting of independent variables obtained from the PCA, were regressed against the dependent variables using OLS regression analysis. In this study, the dependent variables consisted of component factors while the independent variables were the meteorological factors.

The correlation coefficients are similar to the ratio values of pollutant concentrations because the ratio values of air pollutants are less dependent on temporal variations in sources and the meteorological conditions. Therefore, the ratio analysis involving the selected air pollutants is undertaken to examine the pollutants sources. It will

help to identify the major contributors that influence the levels of air pollution on Langkawi. Motor vehicle sources (e.g. cars, buses, motorcycles as well as light- and heavy-duty trucks) are characterized by high concentrations of CO and NO_x while point source emissions (industrial activities) are characterized by high SO₂ and NO_x concentrations [48].

2.6. Trajectory analyses and wind direction

The trajectory analyses used in this study can be divided into two; back trajectory analysis and wind rose analysis. The back trajectory analysis performed using the Hybrid-Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model [49]. The analysis was performed to determine the origin of air masses arriving at Langkawi Island. The analysis can identify the region of Langkawi Island which is most influenced by a given air mass. The input data for running the model analysis was obtained from the Global Data Assimilation System from the National Oceanographic and Atmospheric Administration, NOAA's Air Resources Laboratory. Using input data from between 1999 and 2011, the HYSPLIT model was used to calculate the 3-day (72 h) back trajectories. The trajectories were driven using gridded meteorological data at six-hour time intervals with calculations set for 500 m above ground level to obtain the mean trajectories of the clusters for each season.

In order to determine the distribution of local wind patterns, wind rose analysis was used to complete the study. According to Zelenko et al. [50], wind rose analysis helps to identify the pattern of wind distribution, the distribution period and to determine the direction of the dominant wind. The wind rose analysis was undertaken using the WRPLOT ViewTM software. A complete and detailed wind direction and wind speed dataset from 1999 to 2011 obtained from the DOE was used as an input data to perform the analysis. Ground level wind speed (km h⁻¹) and direction (°) were measured hourly over thirteen years and the result was presented as a wind rose diagram. The diagram described the frequency and the speed of the wind that blows from all directions towards the study area.

3. Results and discussion

3.1. Statistical descriptive analysis

The results from the statistical descriptive analysis consisted of the values of minimum, maximum, mean, first quartile, third quartile, median and standard deviation (Table 1). The PM₁₀ concentration range on Langkawi Island was between 5 µg m⁻³ and 183.2 µg m⁻³ (Table 1). The results show that the 24 h average concentration of PM₁₀ (35.3 µg m⁻³) was below the RMAQG and WHO guidelines, which are 150 and 50 µg m⁻³ respectively. The maximum PM₁₀ concentration was recorded on the 13th August 2005. This year was a year of haze episodes in Malaysia due to

Table 1. Overall Statistical Descriptive Analysis for Langkawi air quality data set (1999–2011). The major air pollutants concentration was compared with the Recommended Malaysian Air Quality Guidelines (RMAQG).

Variables	Minimum	Maximum	Quartile 1	Median	Quartile 3	Mean	Standard deviation	Averaging Time (h)	RMAQG
PM ₁₀ ($\mu\text{g m}^{-3}$)	5.0	183.2	22.9	32.4	44.1	35.3	18.7	24	150
CO (ppb)	10	3830	270	410	633	494	324	1	30,000
O ₃ (ppb)	1.0	84.00	5.50	14.4	24.9	16.7	12.8	1	100
NO ₂ (ppb)	1.0	53.00	2.20	4.40	7.00	5.20	3.60	1	170
SO ₂ (ppb)	1.0	14.00	1.00	1.00	2.00	1.50	0.80	1	130
NO _x (ppb)	1.0	144.0	4.30	9.60	18.8	13.4	12.1	1	-
NO (ppb)	1.0	127.0	2.40	5.60	12.8	9.30	9.70	1	-
Temperature (°C)	19	40.70	24.6	26.5	30.2	27.4	3.60	-	-
Humidity (%)	25	99.00	67.6	80.4	91.6	78.3	14.0	-	-
Wind speed (km h^{-1})	0.0	35.40	2.50	4.90	8.00	5.60	3.80	-	-

uncontrolled forest fires and biomass burning in Sumatra, Indonesia [51, 52]. Haze is most likely to occur during the months of June to September every year, when the weather is dry [53]. The occurrence of haze episodes during this seasons was usually related to trans-boundary smog released by high numbers of biomass burning events in the neighbouring country and influenced by monsoon winds. The south-west monsoon wind, which occur during the months from June to September, will transport the air pollutants from the south-west corner of Southeast Asia and influences the air quality in Malaysia [54].

The average concentration of other gases, CO, O₃, NO₂, and SO₂, in Table 1 were found to be far lower than maximum concentrations suggested by RMAQG. The maximum hourly concentration of CO recorded was 3830 ppb compared to the RMAQG limit level of 30,000 ppb. The O₃ maximum hourly concentration recorded was 84 ppb compared to the RMAQG level of 100 ppb. The average concentration of O₃ was relatively high although the concentration values were below the maximum level recommended by RMAQG. High concentrations of O₃ at this station can be associated with the high intensity of ultraviolet (UV) radiation in the tropical area and the existence of O₃ precursors locally and from trans-boundary sources [55]. The presence of sunlight will induce the reaction of NO_x and volatile organic compounds (VOCs) to form O₃ in the ambient area [56, 57]. Normally, the concentration of O₃ is higher in the middle of the day or early afternoon and dominates the Air Pollutant Index (API) readings in several areas [58]. The maximum concentrations of the other reactive gases, NO₂, NO, NO_x and SO₂ were found to be 53 ppb, 127 ppb, 144 ppb and 14 ppb, respectively. The concentration of NO_x is higher compared to NO and NO₂ values as compound NO_x includes NO and NO₂.

The mean values of air pollutants in this study were compared with other studies made at three different monitoring station locations in Malaysia. There was an urban station located at Petaling Jaya, a background station at Jerantut and a suburban station at Langkawi Island which refers to this study. The PM₁₀, SO₂, NO₂ and CO of this study shows the lowest concentrations among the three locations which are 35.3 µg m⁻³, 1.50 ppb, 5.20 ppb and 494 ppb, respectively. The urban station at Petaling Jaya recorded the value of PM₁₀ at 58.1 µg m⁻³, SO₂ at 8.00 ppb, NO₂ at 33.0 ppb and CO at 1853 ppb. This might be due to the location of Petaling Jaya which is located in Klang Valley that is known as one of the most developed area with rapid urbanization, high population growth, industrial activities and near to major roads [15]. In comparison, Langkawi is a small island that is focused more on tourism development. The island was previously just a quiet fishing community [59]. The background station at Jerantut shows PM₁₀ and SO₂ values that are slightly higher compared to Langkawi Island, which are 37.9 µg m⁻³ and 1.9 ppb respectively. Local biomass burning and agricultural activities might be contributed to the high concentrations of pollutants such PM₁₀ and SO₂ in Jerantut since the location is surrounded with agriculture and residential areas [60]. Low local emission other than rapid

dilution factor due to movement of air with an island may contribute to the low amount of pollutants such as PM_{10} and SO_2 in Langkawi. The O_3 concentration in this study is 17.0 ppb, which is higher compared to Petaling Jaya (14.0 ppb) and Jerantut (12.6 ppb). The higher concentration of O_3 in the island indicated the production of local O_3 that might be from the localized biomass burning and transported pollutant especially the precursor pollutants of biomass burning and anthropogenic activities from nearby area into the island [61].

3.2. Temporal pattern of air pollutants

3.2.1. Diurnal pattern

The diurnal patterns for all observed pollutants on Langkawi Island, along with the meteorological parameters, are presented in Fig. 2. The concentrations of PM_{10} , CO, NO, NO_x , NO_2 and SO_2 all showed similar bimodal distributions while the concentration of O_3 showed a unimodal distribution pattern. Based on the graph in Fig. 2, the concentrations of PM_{10} , CO, SO_2 and NO_x significantly increased between 08:00 and 09:00 and decreased at midday (first peak). Then, the concentrations of PM_{10} , CO, SO_2 and NO_x increased at 17:00 and began to decline after 21:00 (second peak). These two peaks can be attributed to the rush hour traffic on Langkawi Island in the early morning and late evening. The traffic is congested during these periods due to the higher number of vehicles on the road [62]. The second peaks occurred ~3 hours after the evening traffic rush hour indicated to a combination of pollutants emissions and a more stable planetary boundary layer which is characterized by weak turbulence and diffusion, allowing pollutants to accumulate in the layer [63, 64, 65].

The diurnal cycle of NO and NO_2 showed a bimodal pattern, with the first peak between 07:00–09:00 and the second peak between 17:00–21:00 (Fig. 2). However, the concentration of NO and NO_2 reduced significantly in the afternoon. The decreases of NO and NO_2 were correlated with an increase in O_3 during the afternoon. This is due to the photochemical formation of O_3 , where the level of global solar radiation is higher during the afternoon and enhances the production of O_3 since both NO and NO_2 are the main precursors [66]. The unimodal distribution pattern of the O_3 diurnal cycle showed that the peak occurs during the afternoon due to the presence of high levels of UV radiation. The presence of UV radiation is important in the photochemical process of NO_2 releasing atomic oxygen (O). The reaction of atomic oxygen with O_2 will produce O_3 and these two reactions are related to the increase of O_3 to the maximum concentration after the NO_2 peak [56, 67]. NO, which is produced via photochemical processes and emission from vehicles, will react with O_3 to produce NO_2 , the precursor of ozone production [56]. The air pollutant diurnal cycles clearly show that the increase in the number of motor vehicles on the roads greatly influences the air quality on Langkawi Island during the peak hours. The

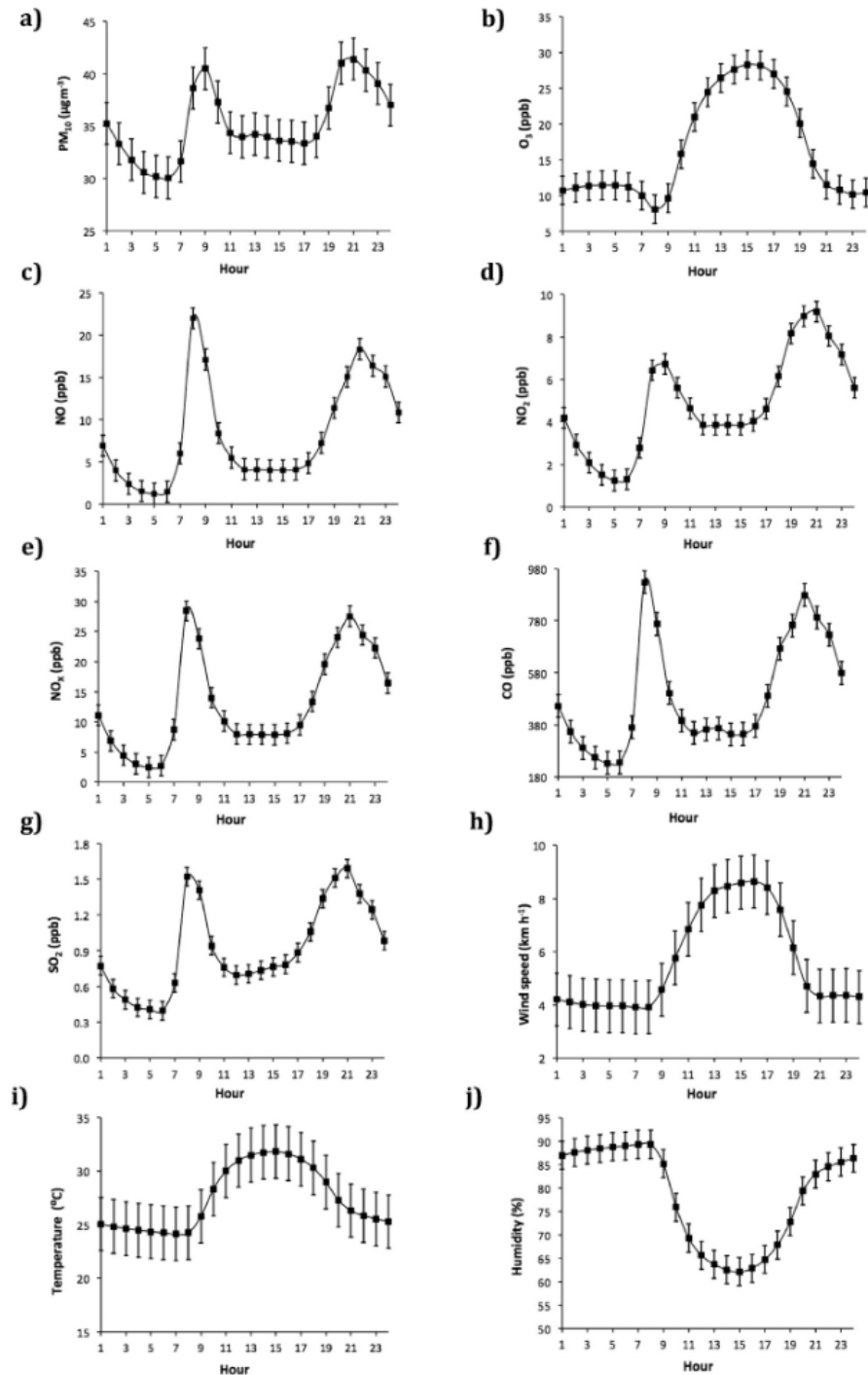


Fig. 2. Diurnal patterns of major air pollutants and meteorological factors recorded in Langkawi between 1999 and 2011. Major air pollutants consist of a) PM₁₀; b) O₃; c) NO; d) NO₂; e) NO_x; f) CO; g) SO₂; while meteorological factors consist of h) Wind speed; i) Temperature; and j) Humidity. The edges of the box plot indicate the 25th and the 75th of the daily concentration of the pollutants.

movement of motor vehicles moving into and out of Kuah, especially in the morning and late afternoon, led to an increase in the concentration of air pollutants on Langkawi Island.

Further investigation on the diurnal pattern of air pollutants on Langkawi Island was undertaken using the cluster analysis known as HACA towards all seven air quality variables (CO , O_3 , PM_{10} , SO_2 , NO , NO_2 and NO_x). The outcomes of the HACA were consistent with the results of the diurnal pattern (Fig. 2). The temporal pattern of air pollutants on Langkawi Island showed three clusters and these were labelled as Group 1, Group 2 and Group 3 (Fig. 3). Group 1 consisted of the hours 01:00, 07:00 to 11:00, and 18:00 to 24:00, and presumably corresponds to the emissions from by early morning and late evening traffic, which led to the increase of PM_{10} , CO , NO , NO_x , NO_2 , and SO_2 concentrations. Group 2 consisted of the hours 02:00 to 06:00 and corresponds to the early morning hours where reduced pollutant concentrations are due to less traffic emissions and anthropogenic activities that contribute to air pollutants in the air. The influence of temperature also becomes a factor in the low air pollutant concentrations on Langkawi Island. Group 3 consisted of the hours 12:00 to 17:00 and corresponds to the high production of O_3 . Group 3 fits the time frame where the levels of UV radiation are higher during afternoon and the presence of higher UV radiation will initiate photochemical reactions and produce more O_3 in the air.

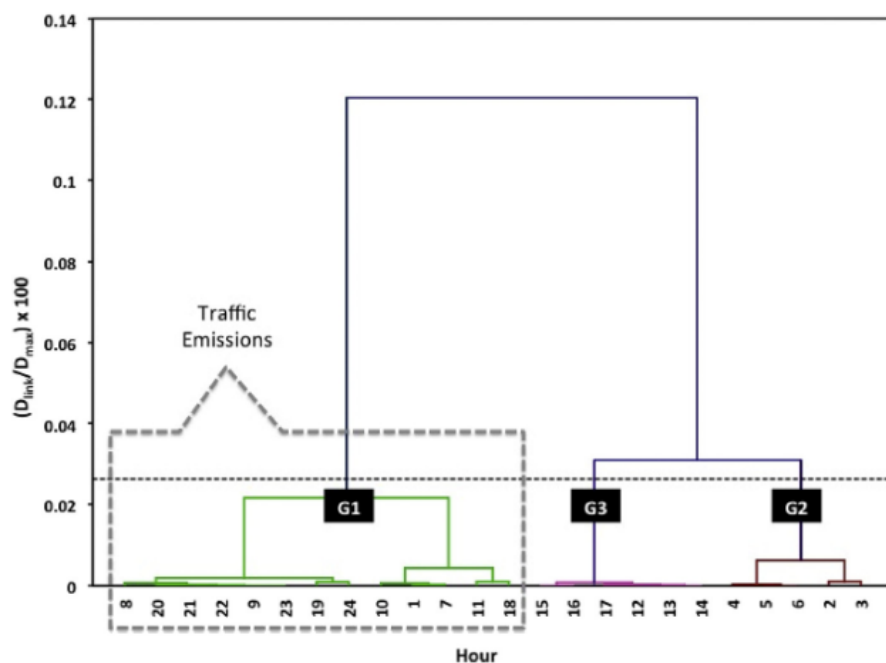


Fig. 3. Dendrogram of diurnal pattern based on yearly average concentrations of all air pollutants in Langkawi (1999–2011). There were three clusters obtained from the HACA analysis labelled as Group 1 (G1), Group 2 (G2) and Group 3 (G3).

3.2.2. Monthly pattern

The data analysis showed that the monthly variations of air pollutants in Langkawi Island were influenced by the monsoons (Fig. 4) with remarkable seasonality. The monthly air pollutant concentrations are illustrated in the box plot graph in Fig. 4. The monthly concentrations for PM₁₀ were recorded at higher concentrations during seasonal monsoon periods: the north-east (NE) monsoon and the south-west (SW)

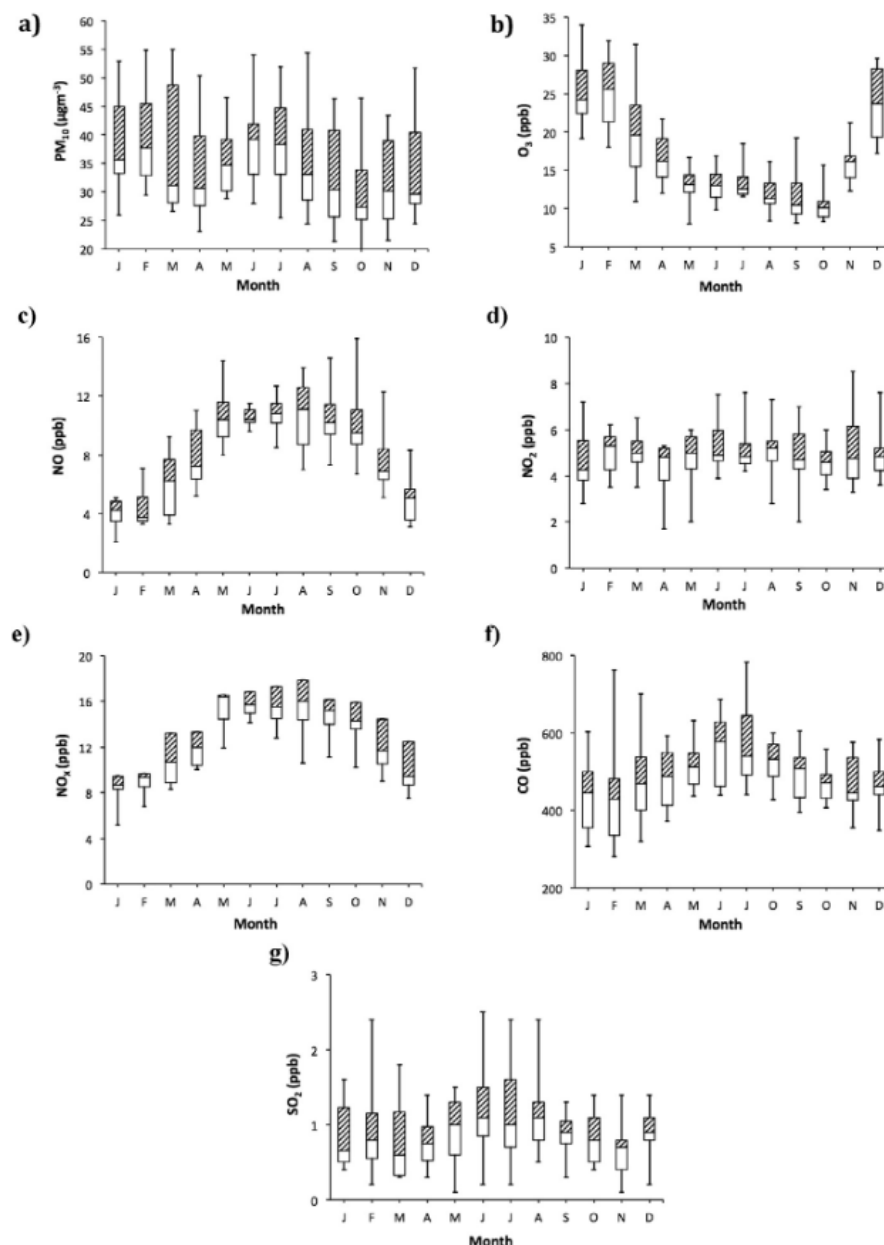


Fig. 4. Monthly patterns of air pollutants recorded in Langkawi between 1999 and 2011 where a) PM₁₀; b) O₃; c) NO; d) NO₂; e) NO_x; f) CO; g) SO₂. The edges of the box plot indicate the 25th and the 75th of the daily concentration of the pollutants.

monsoon (Fig. 4). The NE monsoon occurs between late November and March while the SW monsoon typically starts from late May to September. Fig. 5 presents all the 72 h air mass back trajectories originating from southwest and northeast regions to Langkawi Island, indicating different sources during the SW monsoon and NE monsoon. These higher PM_{10} values were due to the movement of air mass particles typically originating from biomass burning in Sumatra and Indo-China as shown by the majority of the trajectories (Fig. 5). The air masses affected by long-range transport during the NE monsoon originate from the east coast of Indo-China [68]. In addition, agricultural activity from the mainland (northern Peninsular Malaysia) also influenced the concentration of PM_{10} , especially during the NE monsoon, which is shown by the trajectories in Fig. 5b. The high PM_{10} concentrations were influenced by biomass burning in Indo-China which is possibly attributable to the burning of forests and agricultural residue [69, 70]. The wind rose analysis in Fig. 6 shows that the wind came from the northeast and originated from the Indo-China region. The northeast wind was dominant during the NE monsoon and the wind speeds recorded were more than 22 km h^{-1} , which made the transportation of air pollutants long distances from Indo-China possible. The air moves away from the equator in a northerly direction. The strong meridional transport events within the NE monsoon that are able to transport polluted air masses from east Asian pollution to remote parts of equatorial Southeast Asia are caused by the Siberian High pressure system movement and are associated with movement of cold air masses towards southern China and a strengthening of the NE monsoon winds in the South China Sea [71, 72]. According to Streets et al. [70], the highest emissions that contribute to high particulate matter in the air occur in March. This is

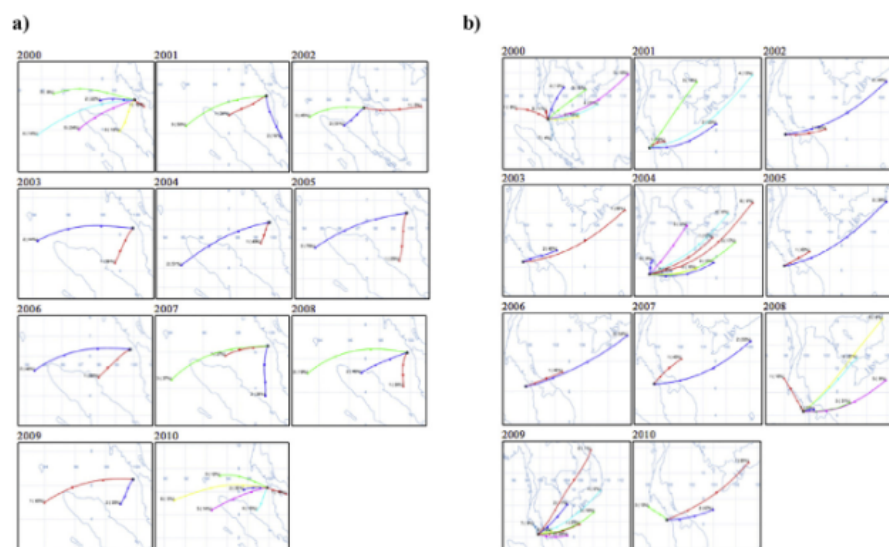
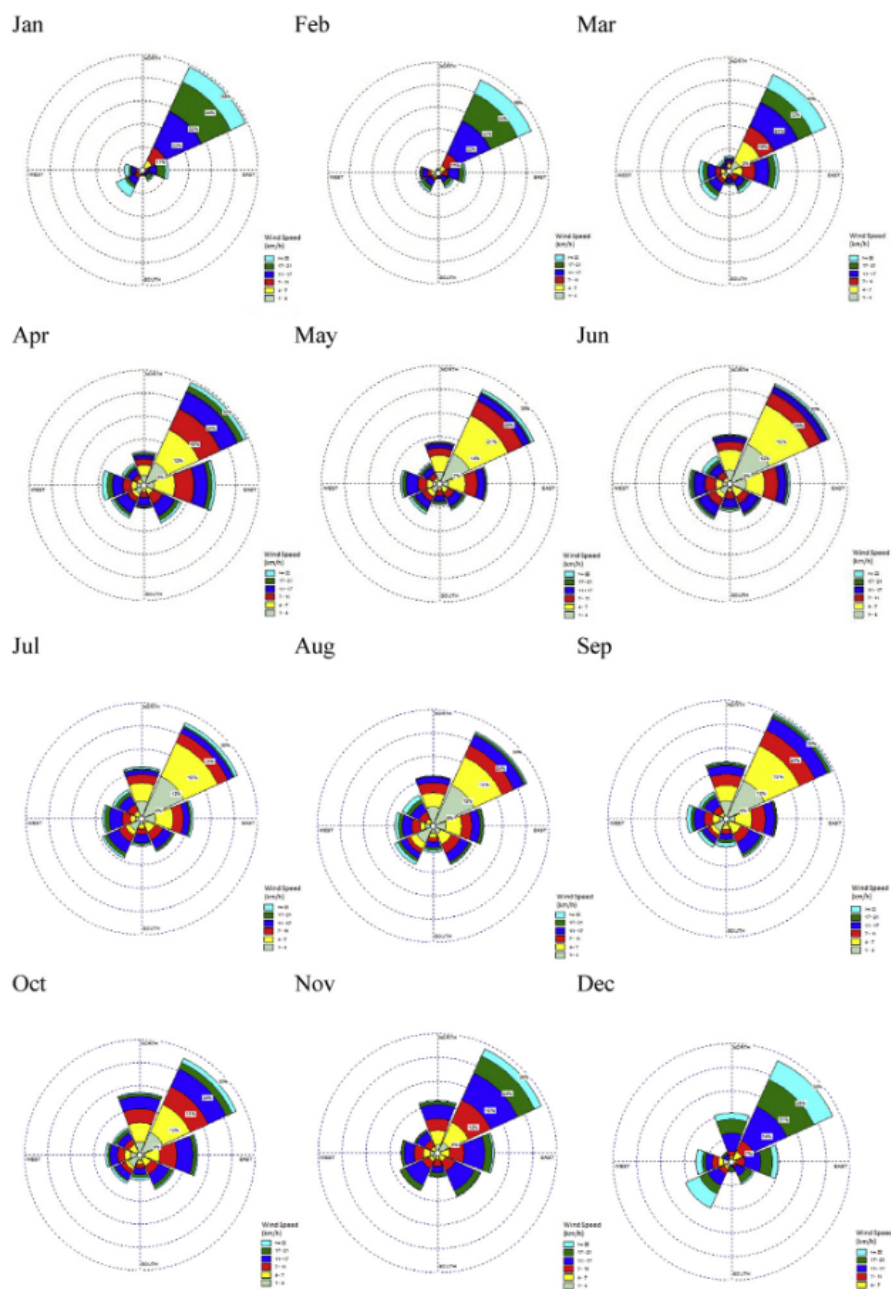


Fig. 5. Mean backward trajectories for the air mass (%) originating in Langkawi Island (marked with an asterisk). Backward trajectories calculated using HYSPLIT model for the individual years from 2000 to 2010 for 500 m altitudes. a) Backward trajectories for SW monsoon (May–Sep). b) Backward trajectories for the NE monsoon (Nov–Mar).



June to August. Air masses travelled shorter distances from Sumatra, Indonesia during the SW monsoon as shown by the trajectories in Fig. 5a. Moreover, the wind rose diagram (Fig. 6) shows that concentrations of CO and SO₂ were high during low-speed north-westerly winds (4–7 km h⁻¹) which causes the air masses to flow from the southwest area with the influence of south-westerly winds. The high concentrations of CO and SO₂ recorded may be due to biomass burning episodes in Sumatra, Indonesia [74]. Biomass burning activities in Sumatra involve peat burning which contributes to CO and SO₂ gas emissions [53, 75]. From November to March, Langkawi is influenced by NE monsoon winds from Indo-China and so the biomass burning episodes usually occurring in Indo-China may influence the concentration of CO and SO₂ on Langkawi Island. However, the CO and SO₂ monthly trend showed less seasonal influence compared to other pollutants (such as PM₁₀ and O₃). The sources of CO and SO₂ were dominated by the emissions from motor vehicles compared to other potential sources including biomass burning and industrial activities like coal mills and charcoal factories [70]. The high concentrations of CO and SO₂ in February are likely to be related to a minimal amount of rainfall received by the island [76, 77]. The slow movement of motor vehicles due to traffic congestion will enhance the concentrations of CO in the atmosphere [78] especially during the minimal amount of rainfall. During December the concentrations of CO and SO₂ remained low although the traffic congestion increase due to school holidays. This situation can be attributed to the high level of rainfall received by the island at the end of the year as high levels of rainfall will washout air pollutants in the atmosphere [79, 80].

The O₃ concentrations were high during NE monsoon, between December to March (Fig. 4). The NE monsoon brings trans-boundary O₃ from Indo-China, the northeast area of Peninsular Malaysia [55]. According to study by Yonemura [81], areas in Peninsular Malaysia are under the influence of air masses from the Northern Hemisphere that are rich in ozone precursor gases [82] because of the movement of the intertropical convergence zone towards the Southern Hemisphere. Due to the positive temperature and minimum water vapour anomaly, the air masses are subject to radiative heating rather than deep convection during the long transport from the Northern Hemisphere, which may lead to the production of ozone during the long period of transport. In addition, Engel-Cox et al. [83] stated that western locations of Peninsular Malaysia appear to have the lowest cloud cover early in the year and highest cloud cover with strong rainstorms in October. The UV radiation values are mostly dependent on the rainfall [84]. The high amount of UV radiation increases the photochemical reactions that produce more O₃ in the air. This further increases the local O₃ concentrations which are already under the influence of from Indo-China during the NE monsoon [55]. The concentrations of O₃ during the SW monsoon season (May–Sep) and inter-monsoon seasons (Apr and Oct) are lower compared to NE monsoon season (Dec–Mar). This situation can be attributed to the NO titration chemistry reaction [85]. The NO titration reaction occurs when

the concentration of NO_x is high. The titration reaction can lead to the destruction of the O_3 and production of NO_2 , which shows an increase when O_3 is reduced [86]. High NO_2 concentration at night time divert the initial step of VOC oxidation by forming nitric acid, which prevent the formation of O_3 as well [87]. In addition, high traffic volume and industrial emissions may contribute to increasing NO_2 in the Langkawi Island [88]. These simultaneous measurements of NO , NO_2 , NO_x and O_3 indicated that the fraction of primary and secondary NO_2 in the Langkawi Island varied depending on mixing and photochemical conditions [89].

3.3. Long term pattern of air pollutants

The overall yearly trends from 1999 to 2011 based on the median concentration of all air pollutants are illustrated in Fig. 7. All air pollutants (CO , O_3 , SO_2 , NO , NO_2 and NO_x) showed a decreasing trend, except for PM_{10} , which showed an increasing trend of $0.24 \mu\text{g m}^{-3}$ per year. The increasing yearly trend of PM_{10} concentration is expected to be influenced by regional factors such as biomass burning and period of dry conditions. The emissions from agricultural activities in northern Peninsular Malaysia and southern Thailand also contributed to the amount of particulate matter in Langkawi's atmosphere, as shown in the backward trajectories in Fig. 5. Paddy straw and sugarcane burning are two major activities which can contribute to PM_{10} concentrations. The presence of El-Niño phenomenon influences the PM_{10} concentrations as it induces dry conditions and therefore encourages burning. The El-Niño phenomenon caused the regional biomass burning from neighbouring countries to become worse and contributed to the high PM_{10} concentrations on Langkawi Island in 2005 [53, 90]. The annual average PM_{10} concentrations (ranging from $20 \mu\text{g m}^{-3}$ to $40 \mu\text{g m}^{-3}$) on Langkawi Island over the thirteen years exceeded the maximum allowable annual standard limit set by the WHO of $20 \mu\text{g m}^{-3}$ but was still under the level recommended by the RMAQG which is $50 \mu\text{g m}^{-3}$. High annual concentrations of particles exceeding that of the WHO annual standard may be affected by regional and local tropical conditions such as wind-blown dust, biomass burning, sea spray as well as anthropogenic sources such as motor vehicles, small industries and domestic burning.

The decreasing trend of O_3 is related to the downward trend of its precursors, such as NO_x and NO , which influence the formation of O_3 with the presence of UV radiation. The negative trend shown by the O_3 concentrations was -0.02 ppb/year. The spike of O_3 concentrations in different years was caused by changing weather conditions such as the rise of global temperature in 2010, which was the second warmest year on record behind 1998 [91]. Although the yearly concentrations of NO_x , NO and NO_2 show descending trends of -0.19 ppb/year, -0.17 ppb/year and -0.04 ppb/year, the yearly trends were influenced by the traffic conditions in Langkawi Island since the traffic volume increased steadily on Langkawi Island. The descending

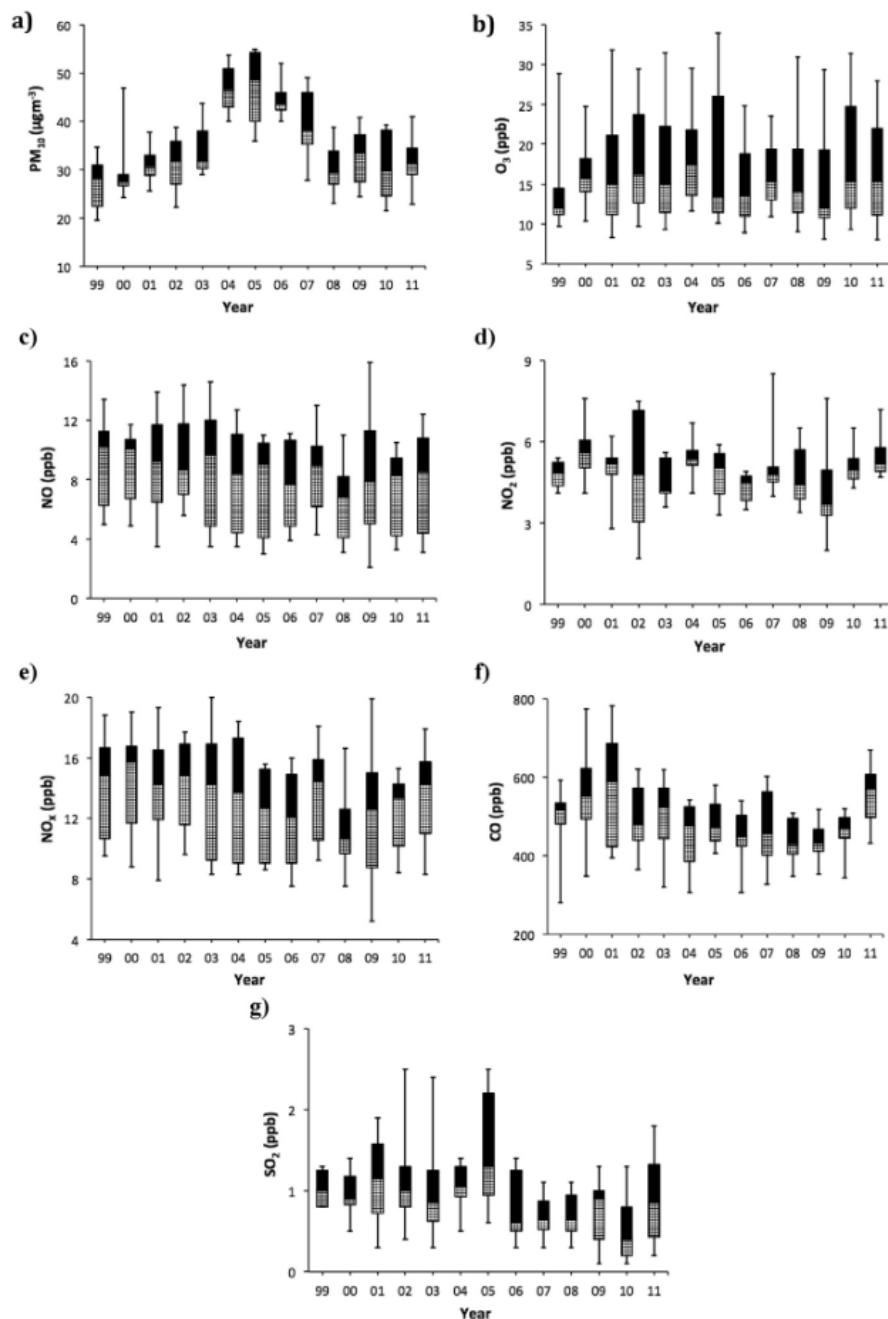


Fig. 7. Yearly pattern of air pollutants in Langkawi (1999–2011) where a) PM₁₀; b) O₃; c) NO; d) NO₂; e) NO_x; f) CO; g) SO₂. The edges of the box plot indicate the 25th and the 75th of the daily concentration of the pollutants.

trends of these pollutants occurred from 1999 to 2008. The trends started to increase from 2009 to 2011, which can be related to the increase in traffic volume on Langkawi Island, since the number of tourist arrivals was increasing. In 2009, Langkawi hosted tourism events such as the Langkawi International Maritime and Aerospace Exhibition (LIMA), the Royal Langkawi Sailing Regatta, the Langkawi Ironman

Triathlon and the Langkawi Water Festival [37]. Increasing tourism events enhanced the number of tourists visiting Langkawi Island.

The negative trend value (-5.55 ppb/year) indicated the decreasing trend of annual CO concentrations (Fig. 7). CO generally originates from incomplete combustion processes from vehicle emissions and open burning. The concentrations of CO were seen to increase from 1999 to 2001 and then started to decline between 2002 and 2008. The increase of CO concentrations during 1999–2001 was attributed to forest fires in the agricultural area that caused the destruction of rubber plantations on Langkawi Island for the purposes of development, especially to develop tourist areas [92]. The decline of CO between 2002 and 2008 was related to lower incomplete combustion from open burning for agricultural activities only, since there were no forest fires for development purposes occurring. Although the increasing trends of CO from 2009 to 2011 were not as high as the trends from years 1999–2001, a rise in CO trends can probably be attributed to the traffic volume in Langkawi Island since number of vehicles increase from 11,274 in 2009–11,899 in 2011 as stated in Road Traffic Volume Report [93]. The annual SO₂ concentrations showed a decreasing trend (-0.03 ppb/year). This is due to the use of fossil fuels with less sulphur content, therefore contributing to the reduction of sulphur emissions to the atmosphere [15]. Although SO₂ has a shorter lifetime, anthropogenic activities such as coal mill factories and traditional charcoal factories at Kilim (located about 20 km from the monitoring station) can contribute to the concentrations of SO₂ in the air. Haze periods that normally happen during the SW monsoon also influenced the concentration of SO₂. The concentration of SO₂ recorded a peak in 2005 due to severe peat burning in Sumatra, Indonesia that led to high emissions of SO₂ to the atmosphere [94].

3.4. Significant ratios indicator for pollution level

The index analysis (ratio) was used in this study to identify the structural changes on the sources of the air pollutants. Only selected air pollutants were involved in this analysis, i.e. CO, SO₂, NO, NO₂ and NO_x, because these pollutants are considered significant variables for mobile and point source emissions [48]. Fig. 8 shows the ratio of CO/NO_x, SO₂/NO_x and NO/NO₂ used in the analysis. Within the thirteen-year observation period, the air quality on Langkawi Island was dominated by the CO/NO_x ratio, which ranged from 28.3 to 43.6 (Fig. 8), while the SO₂/NO_x ratio value was between 0.04 and 0.12, and the NO/NO₂ ratio range was between 1.34 and 2.20. The ratio value of CO/NO_x dominated almost all years except in 1999 and 2009. In 1999 and 2009, the dominant ratio was that of NO/NO₂.

The correlation and linear regression analyses were performed towards each ratio of the selected air pollutants in the index analysis. The correlation analysis shows that CO and NO_x had a positive correlation ($r = 0.12$) and the regression coefficient (R^2) was 0.01. The correlation between SO₂ and NO_x had a positive correlation ($r = 0.10$)

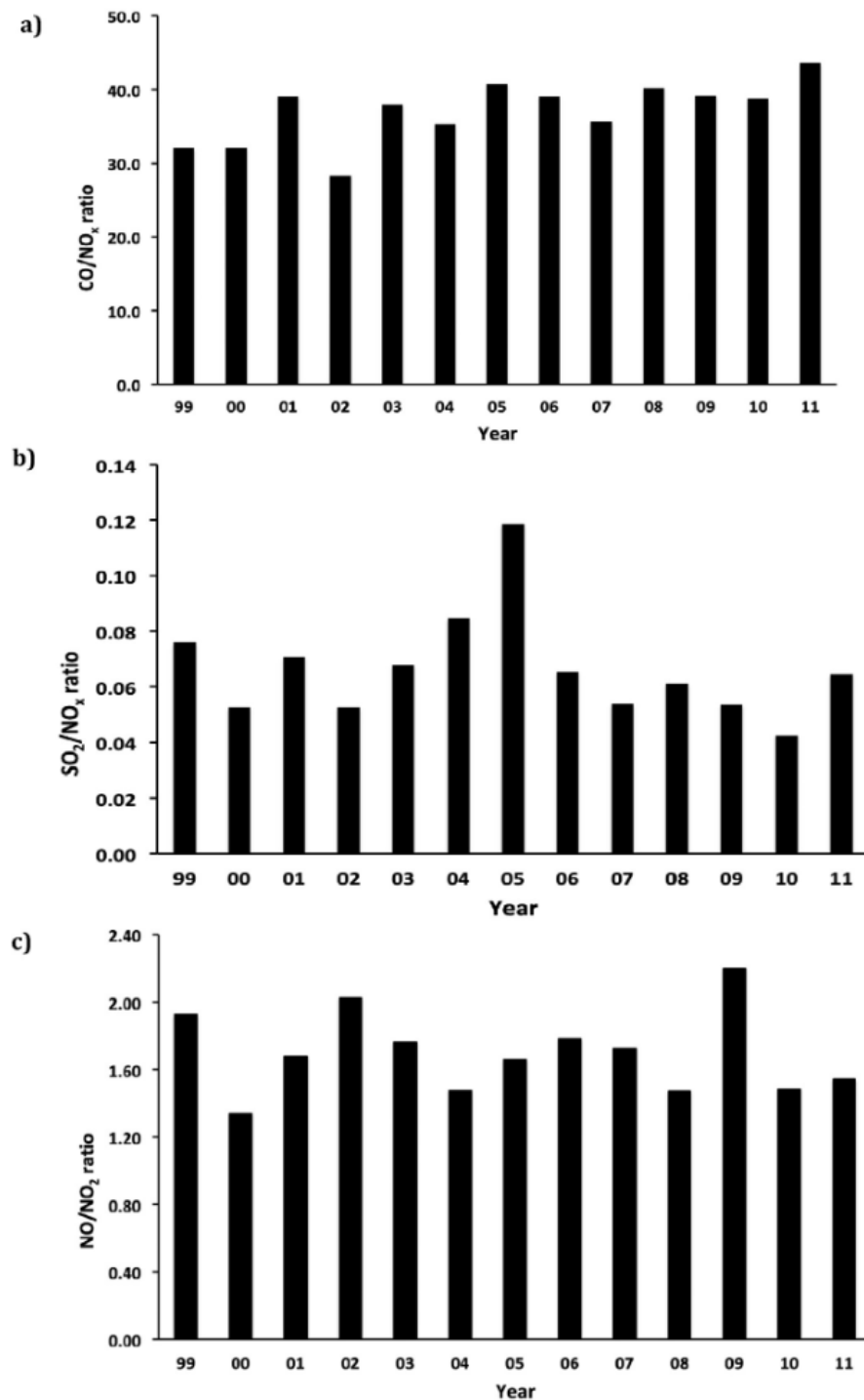


Fig. 8. Hourly averaged values of a) CO/NO_x; b)SO₂/NO_x; and c) NO/NO₂ ratios from 1999–2011.

and the value of R^2 was 0.01. The NO and NO₂ correlation value also showed a positive correlation ($r = 0.23$) and the value of R^2 was 0.05. However, all three ratios were considered have a weak significant correlation value as strong correlation values must be greater than 0.5. This indicates that CO does not have a significant relationship with NO_x. The same applies to the other ratios, SO₂/NO_x and NO/NO₂.

In order to determine the levels of air pollution originating from mobile and point sources, the ratios calculated in this study were compared to other studies. According to Parrish et al. [95] the mobile sources are characterized by a high ratio value of CO/NO_x and a low ratio value of SO₂/NO_x. Point sources are characterized by a low ratio value of SO₂/NO_x and a high ratio value of CO/NO_x. The CO/NO_x ratio is estimated to range from ~ 1 for major point sources to ~ 10 for mobile sources. Hence, from the results obtained, the air quality of Langkawi Island is influenced by major mobile sources, since the ratio of CO/NO_x was 28–44, which exceeds 10, and showed a low ratio of SO₂/NO_x (0.04–0.12) (Fig. 9). Based on a study conducted by Liu [96], the air pollution in an area is considered to be mainly affected by the traffic conditions if the NO/NO₂ ratio value exceeds 2.0. The range of NO/NO₂ ratios in this study was between 1.3 and 2.2. Based on three ratio values, it can be concluded that mobile sources influenced the air quality of Langkawi Island with the weak significant correlation.

3.5. Influence of traffic to air pollutants

In this study, sensitivity analysis using the OAT method was undertaken to examine the level of importance of each air pollutant involved in this study (CO, NO, NO₂,

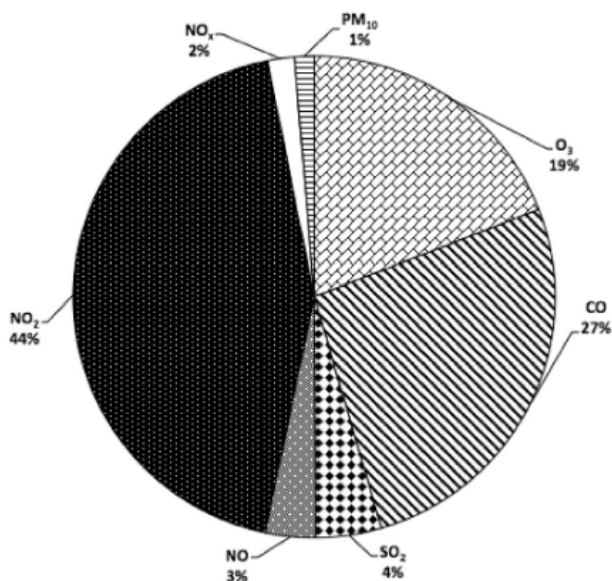


Fig. 9. Pie chart diagram shown a percentage of air pollutant's concentration from the OAT method. The result explained the level of importance for each air pollutant involved in this study (CO, NO, NO₂, NO_x, O₃, PM₁₀ and SO₂) that may be influenced by traffic patterns.

NO_x, O₃, PM₁₀ and SO₂) that may be influence by the traffic patterns on Langkawi Island. The R² values from the OAT method were used to estimate the relationship between traffic patterns and the air pollutant concentrations. The results showed that the seven variables can be classified into three groups based on their level of importance towards the traffic emissions (Fig. 9). The 'least important' group consisted of NO_x, PM₁₀, NO and SO₂. This group showed a distribution range of between 1% and 4% of importance level with respect to the traffic emissions pattern. Next, the 'moderately important' group consisted of O₃ and CO and had a distribution range of between 19% and 27%. The 'most important' group only consisted of NO₂. The motor vehicle emissions were found to influence the NO₂ concentrations the most with 44% of the distribution value. According to Latif et al. [60], CO, non-methane hydrocarbons (NmHC), NO₂ and NO_x are classified as indicators of air pollution from motor vehicle emissions. This outcome clearly shows that the air quality in the study area has been affected by the increasing number of vehicles on the island of Langkawi, since NO₂ and CO were included in the 'moderately important' and 'most important' groups influenced by the traffic conditions.

3.6. Influence of meteorological factors to air pollutants

PCR, which is a combination of the PCA and OLS regression, was used in this study to determine the influence of meteorological factors towards the air pollutants on Langkawi Island [46]. The result of PCA showed that two factors described 80.41% of the cumulative variance (Table 2). The two components that were extracted from the PCA analysis are known as the first factor (F1) and the second factor (F2). The F1 component comprised SO₂, NO and NO_x while the F2 component consisted of PM₁₀. F1 and F2 described the variability of 31.71% and 26.7% of the cumulative dataset variability, respectively. The values of the F1 and F2 components

Table 2. Factor loadings after PCA varimax rotation. The bold values in the table indicate the strong factor loading (>0.75).

Variables	F1	F2
O ₃	0.62	0.66
CO	0.39	0.67
SO ₂	−0.97	0.15
NO	0.98	0.14
NO ₂	0.10	0.69
NO _x	0.96	0.24
PM ₁₀	−0.34	0.84
Eigen values	3.76	1.87
Variance (%)	53.71	26.70
Cumulative of variance (%)	53.71	80.41

were used to run the multiple regression analysis using the OLS method. The standardized regression determination coefficient (R^2) values were used to estimate the relationship between the dependent variables and the independent variables. The dependent variables consisted of component factors (F1 and F2) while the independent variables were the meteorological factors (temperature (T), humidity (H) and wind speed (W)). The equations are illustrated in Eqs. (1) and (2).

$$F1 = 0.103 (H) - 0.702 (T) - 0.230 (W) - 4.807 \quad (1)$$

$$(R^2 = 0.931)$$

$$F2 = 0.409 (T) - 0.155 (W) - 10.367 \quad (2)$$

$$(R^2 = 0.059)$$

The meteorological factor variables influenced about 93% of the F1 component variability with 0.931 for the R^2 value. The meteorological variables affected 5.9% of the F2 component variability with a value of 0.059 for the R^2 . The F1 component indicated a stronger standardized coefficient value for H, T and W compared to the F2 component (Eqs. (1) and (2)). However, the standardized coefficient value for T showed a strong influence on both components F1 and F2. The dominant variable of air pollutants listed in the F1 component included NO and NO_x, which are active precursors that form the secondary air pollutants via photochemical reactions. Photochemical reactions are highly influenced by temperature since dry conditions are typically related to high levels of UV radiation. Hence, they would be strongly influenced by meteorological parameters such as T. The pollutant in F2 (PM₁₀) showed a positive association with T, but a negative association with W. In F1, T had a positive association with SO₂ but a negative association with NO and NO_x. These results showed that the meteorological variables correlated with the changes of the air pollutant concentrations since they influence both the distribution of the air pollutants and the chemical reactions, especially in photochemical processes [35].

4. Conclusion

The results from this study demonstrate that the annual average concentrations of all air pollutants (PM₁₀, O₃, CO, NO, NO₂ and NO_x) on Langkawi Island were below the suggested limits by RMAQG and the WHO. The diurnal patterns showed an increase in all air pollutant concentrations except O₃ during peak hours which are from 07:00 to 08:00 and from 17:00 to 18:00. During peak hours, the traffic conditions around the island, particularly in the area of Kuah town, become congested as these are the start time and end time of typical working hours. The maximum concentration of PM₁₀ was recorded in August 2005 (183.2 µg m⁻³), which was due to the

trans-boundary haze episode from biomass burning in Sumatra, Indonesia. The influence of local and regional emissions are the reasons for the increasing median trend of PM_{10} . Based on the backward trajectory analysis, the monthly trends of air pollutant concentrations were influenced by regional factors such as biomass burning. Aside from haze episode, PM_{10} concentrations did not exceed the suggested limits, although the concentrations were recorded at higher levels during the SW monsoon. O_3 concentrations, with a maximum concentration of 84 ppb, almost reached the level of 100 ppb, the maximum level suggested by the RMAQG. The annual median trends showed that all air pollutants on Langkawi Island had decreasing trends except for PM_{10} .

The long-term patterns of the air quality on Langkawi Island showed that the trends of air quality was influenced by mobile sources due to the high ratio values of CO/NO_x (28.3–43.6) and low ratio values of SO_2/NO_x (0.04–0.12). The air pollution on Langkawi Island was considered to be mainly affected by the traffic conditions since the NO/NO_2 ratio value exceeds 2.0. The result from the OAT regression analysis showed the motor vehicles emissions on Langkawi Island were found to influence the concentrations of NO_2 the most, and secondly CO concentrations. Results from the PCR analysis revealed two components extracted from all seven variables. The F1 component ($R^2 = 0.931$) indicated a stronger standardized coefficient value for meteorological variables compared to the F2 component ($R^2 = 0.059$). The F1 component consisted of SO_2 , NO and NO_x while the F2 component consisted of PM_{10} . The meteorological factors (wind speed, temperature, and humidity) appeared to affect the concentration of air pollutants in the study area as well.

This study suggests, from the concentrations of air pollutants recorded, further air-modelling analysis involving land use studies and emissions inventories on the island is necessary in order to obtain detailed emission source information to achieve a sustainable and clean tourism destination on Langkawi Island. The characteristics of the emission sources obtained by applying ratio analysis and sensitivity methods in this study have provided essential background information on sources in the air quality monitoring study. Development on Langkawi Island needs to be well planned in order to maintain a low level of air pollutants, since motor vehicles contributed significantly to the air pollution in the island. Green technology based management to reduce emissions from waste combustion and local burning will help to maintain lower levels of air pollutants on this island.

Declarations

Author contribution statement

Nor Diana Abdul Halim, Mohd Talib Latif, Fatimah Ahamad, Doreena Dominick, Jing Xiang Chung, Liew Juneng, Md Firoz Khan: Conceived and designed the

experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

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Competing interest statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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